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NATIONAL EMISSIONS INVENTORY OF SOURCES AND EMISSIONS OF SILVER

GCA Corporation

PREPARED FOR
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by

GCA Corporation
GCA Technology Division
Bedford, Massachusetts 01730

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EPA Project Officer: David Anderson

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Office of Air and Water Programs
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May 1973

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1	SILVER MATERIALS FLOW - 1970	5

ABSTRACT

A national inventory of the sources and emissions of the element silver was conducted. The study included the preparation of an overall material flow chart depicting the quantities of silver moving from sources of mining and importation through all processing and reprocessing steps to ultimate use and final disposition. All major sources of silver-containing emissions were identified and their silver emissions into the atmosphere estimated. A regional breakdown of these sources and their emissions was also provided. The physical and chemical nature of the silver-containing emissions was delineated to the extent that information was available, and a methodology was recommended for updating the results of the study every two years.

I. INTRODUCTION

A. PURPOSE AND SCOPE

The Monitoring and Data Analysis Division, Office of Air Quality Planning and Standards of the U.S. Environmental Protection Agency (EPA) has contracted with GCA Technology Division to conduct a national inventory of the sources and emissions of the element silver. The purpose of the study was to define as accurately as possible, based on existing and available published and unpublished information, the levels, nature and sources of silver-containing emissions for defined geographic regions throughout the United States.

The scope of this program is outlined below:

- Develop an overall material flow chart depicting the quantities of silver moving from sources of mining and importation, through all processing and reprocessing steps to ultimate use and final disposition as far as the movements can be traced.

- Identify all major potential silver-containing emissions emitted to the atmosphere from each source. Emission factors and level, and types of air pollution control will also be provided for each of these sources to the extent permitted by available information.

- Define those sources which contribute at least 80% of the total emissions of silver.

- Provide a regional breakdown of these major sources and their emissions.

- Present the nature of the silver-containing emissions for each of these major sources including a delineation of their physical and chemical form and particle size distribution, to the extent that information is available.

- Provide recommendations as to a methodology for updating the results of this study every two years.

B. CONCLUSIONS

1. Material Flow

Based on all available data, 105×10^6 troy ounces (29,000 troy ounces = 1 U.S. short ton) of silver was consumed in the U.S. in 1970. As shown in Figure 1, about 25 percent of this was recovered as old scrap, the remainder being in use or wasted in various ways. The sources of silver included domestic mining and net imports, in about 4:3 proportion. Approximately 160×10^6 ounces underwent primary or secondary refining, of which part was exported and part was recycled as new scrap.

The principal users of silver were the following industries:

Photography ... 30 percent

Contacts and conductors ... 20 percent

Sterling and jewelry ... 20 percent

Brazing alloys, solders, and electroplating ... 20 percent

Other uses ... 10 percent

Certain assumptions made in preparing the above estimates, due to a deficiency of data in certain areas, are explained in detail in Chapter II.

2. Principal Emission Sources

Silver is somewhat unique in that most of the atmospheric emissions of silver are not associated with the silver industry (Chapter II). The principal sources (after emission control, if any) are the iron and steel industry (46 percent), cement industry (25.7 percent), combustion of coal and oil (13.6 percent), and incineration (5.6 percent). One reason for this is that the silver industry is small in comparison with the iron and steel and coal industries. A second reason is the value of silver, which results in close attention to silver processes in which aerosols containing silver are generated.

Within the silver industry, the most concentrated emission sources are primary and secondary smelting and refining (1.1 percent). Although about four times as much silver is emitted in the wearing of

electrical contacts, this emission is spread geographically over a very large number of sources.

3. Regional Emissions

The regions of the U.S. in which most of the estimated emissions of silver occur are Regions 3* and 5, the east central portion of the U.S. The region in which the total emission per square mile is greatest is Region 3, the Pennsylvania-Virginia portion of the U.S. As noted above, these estimates result far more from the geographical distribution and nature of the iron and steel industry than from the silver industry.

4. Nature of Emissions

The principal emissions of silver are believed to be elemental silver, with some simple sulphides, oxides, and halides. This opinion is based on the nature of silver and the characteristics of the high-temperature operations producing the emissions.

Particles containing silver are probably on the order of one micron diameter (MMD). The particles are expected to travel considerable distances (miles) before being deposited.

5. Degree of Control

The overall level of control of silver emission is estimated to have been about 85 percent in 1970. Large sources under relatively poor control included open hearth steel production and municipal incineration. Large sources under relatively good control included blast furnace iron production and commercial incineration.

6. General Comments

The silver content of coal, oil, and/or incineration ash may be sufficient to be of commercial value. Investigation of this might result in added interest in controlling these emissions, and hence, further documentation of this area is recommended.

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* See page 26 for a list of regions.

II. OVERALL U.S. MATERIAL FLOW CHART FOR SILVER

Figure 1* presents a flow diagram depicting the total quantities of silver products moving from sources of mining and importation through the processing and reprocessing steps to ultimate use and final disposition. Each of these sources is discussed below.

A. U.S. PRODUCTION AND ORE PROCESSING (45,006,000 ounces)

Silver is produced mainly as a co-product of base metal ores (66 percent in 1970) and from silver ore (32 percent).[1] In 1970, the separate U.S. sources were (in troy ounces; 29,167 troy ounces = 1 U.S. short ton):

16,023,000 - copper ore processing

14,258,000 - silver ore

11,128,000 - copper-lead, lead-zinc, copper-zinc, and copper-lead-zinc ore processings

2,581,000 - lead ore processing

415,000 - gold ore processing

397,000 - gold-silver ore processing

158,000 - processing of old mine tailings; also tungsten and uranium ore processings

46,000 - zinc ore processing

The Department of Commerce reports the 1970 mining production of silver as 47,483,000 ounces.[2] The above sum and figures appear to be more thoroughly documented, however.

B. IMPORTS AND EXPORTS (Net, 34,671,000 ounces imported)

In 1970 about 40 percent more silver was imported than mined (62,300,000 ounces). Of this, about half was in the form of bullion or refined silver (32,600,000 ounces) and half in forms requiring various types and degrees of refinement (29,700,000 ounces).[1,3]

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*Data in Figure 1 and in this section are left unrounded for purposes of information control. On average, the typical statistic is accurate to within 10 percent, in the opinion of the authors.

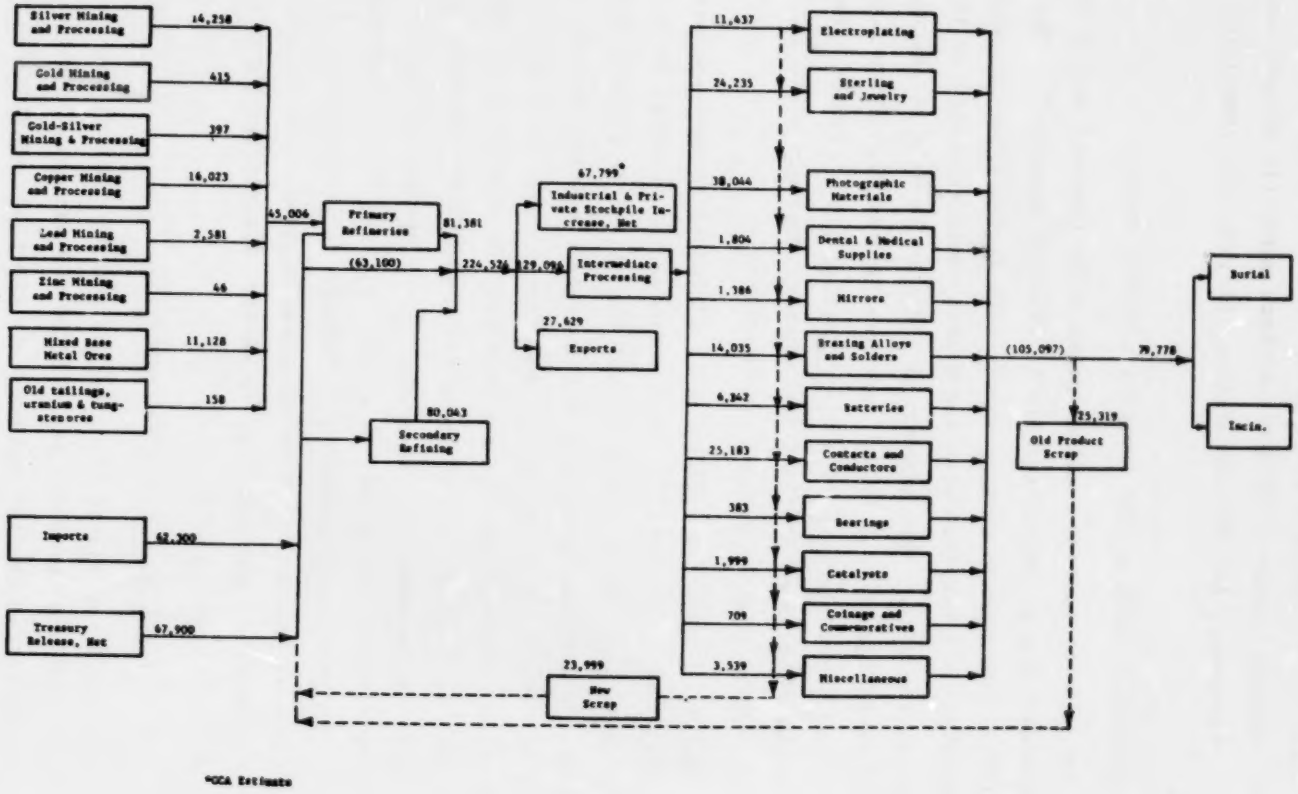

Figure 1 SILVER 1970 MATERIALS FLOW
(THOUSANDS OF TROY OUNCES; 29,000 TROY OUNCES = 1 U.S. SHORT TON)

Total silver exported was roughly one-third of that imported (27,629,000 ounces). Most of this was refined metal (17,240,000 ounces), with waste, sweepings, ore and concentrates (10,378,000) comprising the remainder.(1)

C. STOCKPILE CHANGES (Net Decrease, 101,000 ounces)

About 50 percent more silver entered the domestic supply from Federal sources than was mined in 1970 (67,900,000 ounces).(1) This was chiefly in the form of coinage silver sold by the Treasury, which had accumulated in connection with the recent sharp reduction in the use of silver in coins. This program of large-scale sale of coinage silver was completed in late 1970. Consequently the 1970 stock depletion should be viewed largely as a non-recurring item. (Note: The same reference also gives the 1970 depletion of Treasury stock as 79,000,000 ounces, however without documentation.)

The U.S. industrial stockpile is estimated to have increased by 43,800,000 ounces in 1970.(3) This includes the holdings in U.S. commodity exchanges, but does not include the silver in private hands and in banks. With the cessation of U.S. silver coinage and the sharp rise of the price of silver, private speculative holdings may have increased, although data is not available to support this. We estimate that in fact the sum of industrial, commodity exchange, and private holdings increased by the amount 67,799,000 ounces in 1970. This is approximately the amount of silver released by the Treasury, which was also an unusual event. This release of Treasury silver and the proportionate unusual increase in silver holdings tend to cancel, leaving the imported, produced, and secondary silver flows about the same as in other recent years.

D. SCRAP RECYCLING (49,318,000 ounces)

The total new silver entering the economy (mined, imported, and released from the Treasury) was 175,206,000 ounces. Of this, part entered industrial stockpiles and part was exported, leaving 79,778,000 ounces of new silver for ultimate domestic consumption, after scrap recycling.

The amount of new scrap recycled was 23,999,000 ounces.[1] This is material in relatively pure form requiring a minimum of refinement. The amount of old scrap recycled was 25,319,000 ounces.[1] This includes salvaged products, chemical waste, precipitates, etc., requiring considerable refining. The total scrap recycled (49,318,000 ounces) combined with the new silver to give a total supply of 224,524,000 ounces. After exports and stockpile changes, this left 129,096,000 ounces as the domestic supply. After new scrap, 105,097,000 ounces remained for use in various products. Old scrap was later recovered in the amount of 25,319,000 ounces, leaving 79,778,000 ounces in the form of new products still in use, discarded products not salvaged, and other wasted silver.

E. PRIMARY SMELTING (8.,381,000 ounces)[1]

In addition to the U.S. mining ores and concentrates, U.S. primary refineries also processed 36,375,000 ounces of silver from imported materials, coinage released by the Treasury (containing 90 percent silver), and recycled materials. In general, the type of material to be refined determines whether it requires primary treatment, or whether it can be processed more simply by a secondary refinery.

F. SECONDARY REFINING (80,043,000 ounces)[1]

In addition to old and new scrap, secondary refineries also processed various amounts of imported materials and Treasury coinage silver. There are many such refineries handling a wide variety of scrap and byproducts. The material may pass through several hands and stages of refining before attaining a useful form and purity. The above figure is therefore an estimate, based on the refineries that were considered in the references utilized.

G. CONSUMPTION OF SILVER (129,096,000 ounces)

Purchases of silver were reported as follows:[1]

38,044,000 - photographic materials
25,183,000 - contractors and conductors
24,235,000 - sterling and jewelry

	14,035,000	- brazing alloys and solders
	11,437,000	- electroplating
	6,342,000	- batteries
	1,999,000	- catalysts
	1,804,000	- dental and medical supplies
	1,386,000	- mirrors
	709,000	- government coinage
	383,000	- bearings
	3,539,000	- miscellaneous
Total	129,096,000	ounces

Whether or not these reported purchases include or exclude new scrap is a matter of some uncertainty in the silver industry. The Bureau of Mines and the Department of Commerce both publish "industrial consumption" figures, which however, differ approximately by the amount of new scrap.[20] It is probable that the above figures include new scrap, and that the remainder, 105,097,000 ounces, was actually made into products by the above "consuming" markets. Figure 1 is based on this assumption. After new scrap, 25,319,000 ounces was subsequently salvaged as old scrap from expired products as low-grade wastes, leaving 79,778,000 ounces for ultimate disposal by burial or incineration.

III. SOURCES AND ESTIMATES OF SILVER-CONTAINING EMISSIONS

A. DATA PRESENTATION AND ACCURACY

Table 1 presents a summary of the data from which emissions were estimated for all major potential sources. Each of the six columns comprising this table will be discussed below.

1. Emission Factors

Except where indicated, this gives the pounds of total particulates emitted per ton of production. Such considerations as:

- variations in process conditions among individual plants comprising a source category
- inaccuracies in existing data
- a limited quantity of existing data,

may, however, result in an average emission factor for a source category varying by more than an order of magnitude from the value presented. In recognizing the need to indicate the level of accuracy of these emission factors, a reliability code is presented along with each emission factor value appearing in the table. This reliability code system is described below and is based on the system utilized in EPA Document No. AP-42, "Compilation of Air Pollutant Emission Factors":

A: Excellent

This value is based on field measurements of a large number of sources.

B: Above Average

This value is based on a limited number of field measurements.

C: Average

This value is based on limited data and/or published emission factors where the accuracy is not stated.

D: Below Average

This emission factor is based on engineering estimates made by knowledgeable personnel.

TABLE 1

SOURCES AND ESTIMATES OF SILVER-CONTAINING EMISSIONS

Source	Uncontrolled Particulate Emission Factor		Reliability Code	Production Level (tons/yr)	% Ag in Emissions	Reliability Code	Ag Emissions Before Controls		Estimated Level of Emission Control (%)	Ag Emissions After Controls	
	lb/ton	kg/10^3kg					tons/yr	10^6 oz/yr		tons/yr	10^6 oz/yr
Iron and Steel											
Sinter Process	20 Sinter	10	C	51,000,000	0.01	D	51	1.5	90	5.1	0.15
Blast Furnace	130 Pig Iron	65	C	88,800,000	0.01	D	578	16.8	99	5.8	0.17
Open Hearth	17 Steel	8.5	C	65,800,000	0.05	C	280	8.2	40	168.0	4.90
Basic Oxygen	40 Steel	20	C	48,000,000	0.004	D	38	1.1	99	0.38	0.01
Electric Arc	10 Steel	9	C	16,800,000	0.05	D	42	1.2	78	9.2	0.27
Iron Foundry	20	10	C	22,000,000	0.008	D	18	0.5	75	4.5	0.13
Coal	NA	NA		33,800,000[a]	0.0007	A	236	6.9	82	42.5	1.24
Oil	NA	NA		287,000[a]	0.005	A	14	0.41	0	14.0	0.41
Gasoline, etc.	NA	NA		450,000,000	0.000001[b]	B	4.5	0.13	0	4.5	0.13
Incineration											
Municipal	NA	NA		100,000[a]	0.037	B	37	1.1	37	23.0	0.68
Commercial	--	--		---	---		220	6.5	99	2.2	0.07
Cement	NA	NA		7,790,000[c]	(0.008-0.015) 0.01[e]	B	896[d]	26.0[d]	88	107.0	3.1[d]

TABLE 1 (cont.)

Source	Uncontrolled Particulate Emission Factor		Reliability Code	Production Level (tons/yr)	% Ag in Emissions	Reliability Code	Ag Emissions Before Controls		Estimated Level of Emission Control (%)	Ag Emissions After Controls	
	lb/ton	$kg/10^3 kg$					tons/yr	10^6 oz/yr		tons/yr	10^6 oz/yr
Non-ferrous Metal Processes											
Mining & milling	0.2	0.1	D	45,000,000 oz Ag	NA		0.15	0.0045	0	0.15	0.0045
Primary Smelting	(3-1800)	(1.5-900)									
Copper	1000[d]	500[d]	C	1,765,100 tons Cu	0.035	B	308	9.0	99	3.0	0.09
Other	25	12.5	D	1,460,000	0.175	D	32	0.9	95	1.5	0.045
Secondary Refining											
Copper	NA	NA		210[f]	25 oz Ag/ ton Cu	C	6	0.167	97	0.18	0.0053
Other	32	16	C	2,760 tons Ag	100	-	44	1.28	98.5	0.66	0.019
Consumption											
Brazing & Soldering	1%	---	D	14,000,000 oz Ag/yr	100	-	4.8	0.14	50	2.4	0.070
Electrical contacts	5%	---	D	12,000,000 oz Ag	100	-	21	0.60	0	21.0	0.60
All Other	---	---					1.5	0.043	--	1.5	0.043
Total							2832	82.5	85	416.6	12.1

NA - Not applicable

[a] Uncontrolled emission

[b] % Ag in fuel

[c] Particulate emissions before control

[d] Intermediate value, see text

[e] GCA Estimate

[f] Controlled Cu emissions

2. Level of Production Activity

This column depicts the quantity of material produced (unless otherwise stated) annually. When multiplied by the emission factor, an estimate of the total particulate emissions for that source in pounds per year is obtained.

The values in this column are based on the material flow calculations presented in Section II. Consequently, they have the same accuracy as those material flow values which is estimated at ± 10 percent.

3. Percent Metal in Emissions

The method of analyzing or assaying a dust sample for the amount of metal it contains determines to a large extent the reliability of the data. For example, analytical chemistry techniques for dust containing substantial fractions of metal can be accurate to within a small percentage. On the other hand, optical spectroscopy methods for determining concentrations on the order of parts per million can be inaccurate by a factor of 2. Because of this variability, the reliability codes discussed above for the emission factors are also utilized to estimate the relative accuracy of the percentage values listed in column 3.

4. Level of Emissions Before Control

The values in this column are derived by multiplying the values in columns 1 through 3. The result is converted to tons/year of emissions before control.

5. Estimated Level of Emission Control

The overall effectiveness of control for a source category is based on two factors:

- the portion of the processes which are under control
- the typical degree of control

For example, if 60 percent of vertical roasters have some type of particulate emission control, and these include both scrubbers and

precipitators such that the apparent weighted average efficiency of control is 85 percent, the overall control effectiveness is estimated 60 x 85 = 51 percent.

The accuracy of control efficiency data varies with the degree of control. For a wet scrubber operating at 80 percent efficiency; i.e., passing 20 percent material, the actual emission may safely be assumed to be between 15 and 25 percent because of the relative ease of making determinations at this level. Thus the emissions after control may be assumed to be accurate within $\pm$ 5/20 or 25 percent. On the other hand, for a baghouse reported as being 99 percent efficient, or passing only 1 percent of the material, the actual emission may vary from 0.5 percent to perhaps 2 percent because it is frequently difficult to make low-level measurements with accuracy. In such case, the resulting emission data could be in error by a factor of 2.

Unless otherwise specified, it is assumed that the reported overall level of particulate control applies equally to all silver-containing particles, independent of size, resistance and other important collection parameters. This assumption results in a correct estimate of silver emissions after control when the particulate is chemically homogeneous; i.e., silver is contained in the same concentration in all particles. If however, silver is concentrated in certain particles and in addition the efficiency of the control equipment is not uniform for all particles, then the utilization of an average control level is less valid for calculating silver emissions after control. Data on the preferential control of silver-containing particles is seldom available, but is included in this report when possible.

The accuracy of estimating the level of control for a specific source category is dependent on the quality of available data. The investigators feel that, in general, the level of control data will contribute an accuracy to the resulting emissions estimates within $\pm$ 25 percent.

6. Level of Metal Emissions After Control

The values in this column are derived by multiplying the values in column 4 by the value (100 minus estimated Level of Control).

B. DEVELOPMENT OF EMISSIONS ESTIMATES - 1970

Due to the value of silver, careful control of all processes involving silver is standard practice. Wasting of silver, including airborne particulate, is tolerated only as an economic or practical necessity. Consequently, the emissions containing silver, listed in Table 1, tend to be smaller in proportion, as well as in magnitude, than have been identified for the other metals in this review. The largest silver emissions (iron and steel processing; coal combustion; incineration) comprising about 85 percent of all silver emission, are not a part of the silver industry, but are the inadvertent results of other practices. Data pertaining to these largest emissions are limited for two reasons: The silver industry has not been interested in them as economic sources of silver; and silver has thus far had a low priority as an air pollutant.

1. Iron and Steel Production

Very little information has been found giving typical Ag contents of the emissions from the common iron and steel manufacturing processes. Orban, et al.[4] found a "trace" of Ag_2O in the emission from a Bessemer converter, with an apparent resolution of his tests of about 0.02 percent. Hammond, et al.[5] reports 0.05 percent Ag in the particulate discharge from an open hearth furnace, based on "qualitative" spectrographic analysis with an apparent test resolution of 0.01 percent. The same authors also give an average of 0.0091 percent Ag in samples from a baghouse serving a gray iron cupola furnace, based on a qualitative analysis. No other directly useful data was found.

Additional perspective is gained indirectly, however, in other ways. The average concentration of Ag in the earth is 0.000002 percent;[6] and in coal ash, 0.0007 percent (see below). Ag

tends to occur naturally with some other metals, zinc in particular, and also copper, aluminum, and manganese. These metals bracket the melting and vaporizing temperatures of Ag (960°C and 1950°C, respectively, see Section V). Because they are usually emitted in larger concentrations, they are more easily measured and more often reported. Because of their similarity to silver's thermal characteristics, the ratio of silver to these metals may tend to be preserved in emissions from high temperature processes. In those iron and steel emissions for which data is available, the ratio of silver to zinc appears to be of the order of 0.003, and of silver to copper, 0.10. Data on zinc and copper emissions from all processes listed in Table 1 are generally available.[7,8] Using these known emissions and the apparent ratios, plus the actual silver emission data cited above, estimates of silver emission were made as given in the table. They vary from 0.004 to 0.05 percent.

Table 1 indicates that the iron ore blast furnace generates one of the largest quantities of silver, equal in magnitude to one-third of all the silver mined in the U.S. annually. Because the blast furnace is well controlled, however, the final silver emission is not outstanding. The open hearth furnace, on the other hand, because of a lower level of particulate control, emits approximately 37 percent of all Ag emissions in the U.S. This quantity is equivalent to about 10 percent of the silver mined domestically.

Iron and steel production as a whole is estimated to contribute 58 percent of the silver emission. It is noteworthy that the silver content of these emissions is estimated to be similar to those of copper and lead refinery emissions. The latter are collected when possible and processed for their valuable metal contents. A silver content of 0.01 percent is equivalent to 2.9 ounces/ton, a marginal concentration in ore form, but perhaps more attractive in fume form. Current prices of refined silver are approximately $2.00/ounce.

2. Coal and Oil Combustion

Silver and coal sometimes occur together in nature. For example, in Utah in the 1870's, thin scales of silver were found along with small deposits of minable lignite in sandstone. Evidently almost any concentration of Ag in coal could be encountered depending on the size of the coal sample and its source. Abernathy and Gibson,[9] in a survey of trace metals in coal, cites reports of maximum silver contents of 0.0001 to 0.0084 percent in the ash of a variety of western and West Virginia coals. The geometric mean is about 0.0009 percent. Brown[6] gives the maximum silver content of coal ash as 5 to 10 ppm, equivalent to 0.0005 to 0.0010 percent silver. Accordingly, the average of 0.0007 percent is used in Table 1

Table 1 estimates that slightly less silver emission is developed from coal combustion than from the open hearth process. Furthermore, coal power plants are controlled about three times as well as the open hearth process (82 percent[32] versus 40 percent control). Thus, controlled silver emissions from coal burning contributes about 15 percent of the U.S. silver emissions.

The above references indicate that the ratio of silver to copper in both the earth, and in coal, is about 0.002; and of silver to zinc, about 0.0005. These ratios may be useful in arriving at order of magnitude estimates of the silver content of other samples of coal ash that have been tested for copper or zinc, but not for silver.

Gerstle, Cuffe, et al.[10,11] report the emissions of numerous other trace metals in coal ash, but not silver. Their data give the efficiency of an electrostatic precipitator on each of the metals. The copper-zinc ratio in the ash was reduced by one-half in passing through the precipitator. This suggests that silver may also be differentially controlled by the equipment in use. We have used the average 82 percent level of control, however.

The combustion temperatures of coal are in the vicinity of 1700°C, approaching the vaporization temperature of silver. Thus,

part of the silver in the coal may be vaporized within the firebox. It is probable that this silver vapor condenses into the ash as the combustion products cool. Thus, we have assumed that the ash assay, as reported above, includes essentially all of the silver emitted by the process. The same assumption applies to iron and steel and other high temperature process emissions in Table 1.

Analysis of the ash from fuel oil would be expected to be similar to that from coal, in the sense that both are fossil fuels. Comparisons of typical analyses of each show similar concentrations of copper and zinc. Levy, et al.[21] report a concentration of 0.0018 percent silver in oil ash (optical emissions spectroscopy), moderately higher than the coal ash assay. On the other hand, Walsh[12] reports a concentration of 0.03 percent silver in oil ash (spectrographic analysis). Trace metal content of oil is known to vary by factors of 25 or more, depending upon the type of oil.[13] All factors considered, an average of 0.005 percent is used in Table 1. Oil combustion emissions are uncontrolled, resulting in a level of silver emissions from fuel oil combustion (0.41×10^6 ounces) which is about one-third the level emitted from coal firing.

3. Gasoline and Other Light Fuels

Analyses of premium and low-lead gasolines indicated silver concentrations ranging from less than 0.001 to 0.4 ppm with apparently reliable data of 0.002 ppm (neutron activation), 0.02 ppm (spark source mass spectroscopy), and 0.05 ppm (spark source mass spectroscopy), being reported.[23] An average content of 0.01 ppm is assumed. The silver contents of other light fuels were not available, and were assumed to be the same as for gasoline. It is further assumed that the silver in these fuels is entirely emitted to the atmosphere. Including kerosene, light distillates, and jet fuels with gasoline, 3.4×10^9 barrels or about 450×10^6 tons were burned in 1970. The resulting emission estimate of 0.13×10^6 ounces of silver is shown in Table 1.

4. Incineration

Assays of samples of fly ash from refuse combustion have indicated a silver content of 2 to 9 ounces per ton of ash (equivalent to 0.006 to 0.028 percent Ag).[14] Another source reports a content of 19 ounces per ton (0.059 percent Ag).[15]

An estimated average value of 12 ounces per ton of fly-ash, or 0.037 percent Ag, is used in Table 1. Note that at this rate, the recovery value of the silver is about 14 times greater than the cost of disposal of the ash that was recently estimated to be $1.50/ton.[14] The net emission of 0.68×10^6 ounces is the third highest emission in the table.

Recently, special purpose commercial incinerators have been developed specifically to recover the silver used in photographic materials.[14] Of the 38×10^6 ounces of silver used annually by the industry, it is estimated that 14×10^6 ounces is recycled as precipitates from photographic processing solutions. Of the remainder, it is estimated that 6.5×10^6 ounces is processed by the special incinerators, the rest being burned in municipal incinerators or wasted in various other forms. The special incinerators are reported to recover at least 99 percent of the silver, resulting in a net emission of only 0.07×10^6 ounces from commercial incinerating operations.

5. Cement

Assays of dust from three cement plant clinker coolers indicated 0.015 percent silver (spectrographic analysis) according to a review of EPA data.[22] Assays of dust from three air separators indicated 0.008 percent silver, also from EPA data. Based on these percentages (0.008 to 0.015 percent), the range of silver emissions is 75 to 140 tons/year after 88 percent control with an intermediate value of 107 tons/year in this survey. The resulting emission estimate of 0.32×10^6 ounces is only about 3 percent of the total U.S. silver emission.

6. Production of Non-Ferrous Metals

a. Mining

Mining and milling operations are estimated to generate 200 pounds of emissions per 1,000 tons of production, for each of several non-ferrous metals.[17,18] If one assumes that the concentration of silver in the emitted dust is the same as in the ore being processed, then 200 pounds of silver emissions are generated for each 1,000 tons of silver produced. In 1970, 45×10^6 ounces of silver were produced. Thus the estimated emission of silver from all non-ferrous mining and milling operations generating silver as a co-product is 4,500 ounces. This is after particulate control, which is minimal.

b. Primary Smelting

In the case of copper smelting, Davis reports 10.1 ounces of silver per ton of emitted dust (0.035 percent Ag), and 10 pounds of emitted dust per ton of copper produced, after control by an electrostatic precipitator at about 97 percent efficiency.[17] This measurement was made at the stack, where effluents from various operations had been combined, notably, roasting, reverberatory furnace smelting, and converting. This indicates an effective before-control generation of 333 pounds of dust per ton of copper. In other plants, it was reported that emission factors of 704 pounds of dust per ton of product was generated in smelting before 99 percent control; 1833 pounds per ton of product in smelting before 99 percent control; and 3.1 pounds per ton of product in unspecified processes before control.[24] Using an average 1,000 pounds per ton generated, with a copper production of 1,765,000 tons in 1970 and a control efficiency of 99 percent, results in approximately 90,000 ounces of emission as shown in Table 1.

In the smelting of other ores, an estimated 50 percent of the 1970 U.S. silver production was derived through lead, zinc, lead-zinc, and silver ores.[1] The silver content in these ores averages about 8.7 ounces per ton, whereas in copper ore the content is only about 0.07 ounces per ton (1963 data).[19] The ore is first

concentrated by flotation in which little aerosol is generated. Afterward, there is still a much higher proportion of silver along with these other metals than with copper. A limited amount of emission data is available.[24] Sintering and sinter crushing at a lead smelter is reported to generate 0.3 pounds of dust per ton of product before an average control of 92 percent. For a lead blast furnace and a reverberatory furnace, emissions are reported to be 20 and 36 pounds per ton, respectively, before 95 percent control. Zinc smelting in general is reported to emit 7.0 pounds per ton before an average 90 percent control.

In none of these cases is the silver content of the emitted particulate reported. Since the data base is insufficient for a complete computation of silver emission, the following estimates are made: particulate emission averages 25 pounds per ton, on a base of 1,460,000 tons of heavy metal produced in 1970; the average assay of the particulate is five times greater than for copper due to the higher proportion of silver present; and control efficiences average 95 percent. As a result, 45,000 ounces are estimated to be emitted after control.

Less than 1 percent of the silver produced in the U.S. in 1970 was produced by the processes of amalgamation and cyanidation. In the former process, after crushing, washing and drying of the ore, it is mixed with mercury and then heated in a retort furnace. In the latter process, the crushed ore is leached, after occasional roasting, and the silver is precipitated from the leachate before entering a reverberatory furnace. It is estimated that 1 percent of the silver produced by these two processes escapes as aerosol, or 1,200 ounces.

c. Secondary Refining

The refining of scrap metals other than silver scrap is believed to be relatively free of silver emissions since most scrap has a low silver content. One possible exception is copper scrap from electrical products, which contains considerable silver in

the form of electrical contacts and conductors. The 1970 consumption of silver for contacts and conductors was 25×10^6 ounces; and the consumption of copper by the electrical industry, about 50 percent of the U.S. copper consumption was 1.0×10^6 tons. The resulting ratio is 25 ounces of silver per ton of copper. Emissions of copper from secondary copper refining after controls have been estimated at 210 tons per year.[17] If the consumption ratio applies to these emissions, then it is estimated that up to 5,250 ounces of silver is emitted during secondary copper refining. At 97 percent control efficiency, this would be 167,000 ounces before control.

Emissions from the secondary refining of lead in blast furnaces are reported in two cases.[22] One furnace controlled by scrubber (estimated efficiency 90 percent) produced 1.14 pounds per ton of particulate after control, containing approximately 30 ppm of silver. The second furnace controlled by cyclone and fabric filter (estimated efficiency 99 percent) produced 0.17 pounds per ton of particulate after control containing about 170 ppm of silver. The computed pre-control emission factors average about 15 pounds per ton, and the post control assays average 0.01 percent. Only about 75,000 tons of scrap lead were refined in 1970, resulting in a negligible 0.05 tons of silver generated before control.

Other than the commercial incineration of photographic materials included above, silver scrap is generally refined by melting followed by electrolytic refining. A typical reverberatory furnace may emit 30 pounds per ton of metal, and an electrolytic cell, 2 pounds per ton,[17] for a total emission of 32 pounds per ton. These emissions are controlled at an estimated 98.5 percent. Since only 80,043,000 ounces of silver underwent secondary refining in 1970, the emission is estimated to have been 1,280,000 ounces before control, and 19,200 ounces after control.

7. Intermediate Silver Products

Practically all of the 129,096,000 ounces of silver consumed by the various markets first passes through intermediate hands,

where it is processed in various ways.[20] The metal, in various degrees of purity, may be alloyed; shaped into wire, sheet, powder, etc; or converted chemically or electrochemically. Few, if any, of these processes emit significant amounts of silver, because relatively few firms convert most of the silver. Also, these processes usually involve limited amounts of silver and are closely supervised.

In forming powdered silver, a jet of molten metal is combined with a high velocity air jet. From 40 to 90 percent of the particles thus formed are 50 microns or under. It is unlikely that any appreciable number of particles 5 microns or smaller are produced by this process. Since larger particles will be well controlled, it is assumed that the resulting emission of silver is negligible.

Nearly all silver for photographic consumption is first converted to silver nitrate by dissolving the metal in hot nitric acid in an enclosed vessel. The fumes that escape this vigorous reaction are scrubbed to control the nitrogen oxides produced. The literature on this process is preoccupied with gaseous control problems rather than silver loss. It is unlikely that any appreciable quantity of silver escapes. Similarly, electrochemical conversion of silver into silver sulphate may involve the boiling of hydrogen gas, with possible minor emission of silver. Silver sulphate, chloride, cyanide, and oxides are produced in relatively small quantities compared to silver nitrate.

Little aerosol is emitted in shaping silver. The various steps may include weighing and charging a crucible for melting; poring and casting into molds; rolling, slitting, drawing. As a percentage of all U.S. emission of silver, these sources are estimated to be negligible.

8. Consumption of Silver

a. Electrical Contacts

About one-half of the 25,183,000 ounces of silver used for contacts and conductors, or 12,000,000 ounces is estimated to

be used annually for solid (cast or machined) and composite (laminar) contacts. Although silver is used because it withstands sparking and mechanical wear well, it is evident that the contacts do nevertheless undergo wear. It is assumed that the contacts are designed to last the life of the rest of the electrical equipment, which is typically about 20 years. That is, it is estimated that 5 percent of the silver consumed annually is worn away in the form of aerosol. This results in 600,000 ounces of emission per year. This is spread over a very large number of locations, as most appliances, automobiles, telephones, etc. use some silver contacts.

b. Brazing and Soldering

About 14,000,000 ounces of silver are consumed by brazing. Although the production of these alloys is relatively non-emitting and easily controlled, the brazing process emits a considerable amount of fume. An estimated 1 percent of silver in these alloys is emitted, the rest of the metal remaining at the point of joining. Most of the brazing is done in specialized locations (air conditioning and refrigeration is estimated to consume 20 percent; automotive and aircraft, 30 percent) which permits these emissions to be better controlled than others (plumbing and heating, and electrical appliance brazing tend to be non-concentrated). Assuming that emissions from brazing and soldering are 50 percent controlled, the net emission estimate is 70,000 ounces of silver.

9. Other Sources

GCA estimates that these sources contribute 43,000 ounces of silver emissions, or about one-half percent of the total. The following minor sources have been considered:

Silver Consumption	Emission Estimates (ounces)
Conductors, spray painting	10,000
Bearings and lubricants, loss of flake silver lubricant in use	2,000
Mirrors, spraying of solutions	1,500
Batteries, sintering, electrolytic processes, chemical losses	900

Silver Consumption	Emission Estimates (ounces)
Electroplating, chemical and liquid spills	250
Catalysts, preparation, regeneration and erosion	250
Sterling and jewelry, buffing and polishing	200
Brazing alloys and solder, production	150
Medicine and dentistry, preparation of materials	100
Miscellaneous Sources	
Non-metallic mineral operations emitting dust with trace silver	20,000
Forestry and agricultural burning of unwanted waste containing trace silver (Note: Forest fires not included)	7,500
Cloud seeding	negligible
Wear of coinage	negligible
TOTAL	42,850

C. SUMMARY OF PRINCIPAL EMISSIONS

Table 2 summarizes the largest 1970 emissions of silver, as the estimates were developed in Table 1. These are grouped into industrial sources, those directly associated with the silver industry, and inadvertent sources, those having little to do with the silver industry. In the case of silver, the latter category is the larger.

TABLE 2

PRINCIPAL SOURCES OF SILVER-CONTAINING EMISSIONS - 1970

Source	Ounces/year	Tons/year	Percent*
Inadvertent Sources			
Iron and steel production	5,630,000	193.0	46.3
Cement production	3,100,000	107.0	25.7
Coal burning	1,240,000	42.5	10.2
Municipal incineration	680,000	23.3	5.6
Oil burning	410,000	14.0	3.4
Silver Industry Sources			
Electrical contacts	600,00	20.6	4.9
Primary smelting	133,000	4.5	1.1
Percent of U.S. Total			97.2

*Percent of Table 1 total, 12,100,000 ounces/year.

IV. REGIONAL DISTRIBUTION OF PRINCIPAL SOURCES AND EMISSIONS

For purposes of showing geographical distribution, the U.S. was divided into ten regions identical to the Regional Branches of EPA:

Region	States
I	Connecticut, Maine, Massachusetts, New Hampshire Rhode Island, Vermont
II	New Jersey, New York, Puerto Rico, Virgin Islands
III	Delaware, Maryland, Pennsylvania, Virginia, West Virginia, District of Columbia
IV	Alabama, Florida, Georgia, Kentucky, Mississippi, North Carolina, South Carolina, Tennessee
V	Illinois, Indiana, Michigan, Minnesota, Ohio, Wisconsin
VI	Arkansas, Louisiana, New Mexico, Oklahoma, Texas
VII	Iowa, Kansas, Missouri, Nebraska
VIII	Colorado, Montana, North Dakota, South Dakota, Utah, Wyoming
IX	Arizona, California, Nevada, Hawaii and the South Pacific
X	Alaska, Idaho, Oregon, Washington

Emissions from the principal sources listed in Table 2 are distributed among these ten regions as shown in Table 3. Also, the number of plants producing the emissions is shown in the table when such information is available.

The accuracy of the distributions by region varies with the category. The number of plants per category ranged from one to several thousand in this study. When the number of plants was less than about 100, an attempt was made to identify each plant and plant location, and include it in one of the ten regions. When production or capacity figures for these plants were available, total production or capacity for each region was computed, and the U.S. emission estimate for that category was distributed by region accordingly. When production or capacity figures were not available, the emission was distributed by the number of plants in each region. If the number of plants was very small or

TABLE 3

GEOGRAPHICAL DISTRIBUTION OF EMISSION SOURCE AND EMISSIONS

Emission Source	EPA Regions										Totals	Units	Reference
	1	2	3	4	5	6	7	8	9	10			
Inadvertent Sources													
Iron and Steel Open Hearth	3.7	3.7	65.7	7.3	62.3	7.3	0	7.3	11.0	0	168	(TPY)	
	1	1	18	2	17	2	0	2	3	0	46	(O.H. Plants	25
Other	1.4	2.0	6.7	2.4	8.8	1.2	0.3	0.1	1.7	0.4	25	(TPY)	
	23	33	111	39	146	20	5	2	29	7	415	(All types Plants)	25
Coal Burning	0.3	2.4	9.3	9.1	17.6	0.6	1.7	1.4	0.3	0.1	42.5	(TPY)	
	0.7	5.7	21.8	21.1	41.3	1.4	4.1	3.3	0.7	0.3	100	(Percent Coal)	1
Municipal Incineration	3.8	5.5	4.1	2.7	5.5	0.8	0.3	0.1	0.1	0	23	(TPY)	
	16.6	24.2	17.7	11.8	24.0	3.5	1.4	0.3	0.5	0	100	(Percent Capacity)	26
Oil Burning	2.5	4.1	2.1	1.3	1.1	0.5	0.1	0.3	1.6	0.4	14	(TPY)	
	18.0	29.1	15.0	9.6	7.6	3.7	0.9	1.8	11.6	2.7	100	(Percent Oil)	27
Cement Production	0.6	7.7	16.8	15.8	17.9	15.8	11.8	5.3	11.2	4.1	107	(TPY)	
	1	13	28	27	30	27	20	9	19	7	181	(Plants)	28
Silver Industry Sources													
Electrical Contacts	1.2	2.6	2.4	3.2	4.4	2.0	1.1	0.5	2.3	0.7	20.6	(TPY)	
	5.8	12.5	11.5	15.7	21.6	10.0	5.5	2.7	11.3	3.2	100	(Percent Population)	29
Primary Smelting	0	0.2	0.4	0.2	0.4	1.1	0.3	0.7	1.0	0.4	4.5	(TPY)	
	0	2	4	2	4	11	3	7	10	4	47	(Plants)	30,20
Totals	13.0	28.2	107.5	42.0	118.0	29.3	15.6	15.7	29.2	6.1	405	(TPY)	

there was reason to believe that certain plants were larger or produced more emission, distributions were weighted accordingly.

On the other hand, when the estimated number of plants was greater than about 100, and the distribution of plants was not known, the regional breakdown was made on a different basis, such as population, geographical area, or shipments reported, as most appropriate for that category. Whether the distribution was by plant size, number of plants, or another statistic, the distribution is believed to be accurate to within 10 percent in most cases.

The emissions distributions in Table 3 are made on the following basis:

Inadvertent Sources

Iron and Steel Production:	Proportional to the percentage of U.S. plants in each region
Coal Burning:	Proportional to coal shipments, by state of destination
Municipal Incineration:	Proportional to incinerator capacity, by state
Oil Burning:	Proportional to percentage of residual oil burned
Cement Production:	Proportional to number of plants, by state

Silver Industry Sources

Electrical Contacts:	Proportional to population; i.e., consumption of equipment containing contacts
Primary Smelting:	Proportional to number of smelters of all types, by state

The largest emissions of silver in 1970 are estimated to have been in Regions 3 and 5 (the east central portion of the U.S.). This conclusion is strongly influenced by the production of steel by the open hearth process in these regions, and by the concentration of silver reported in open hearth fumes. Since this concentration report was apparently based on a single measurement of questionable reliability, it is urged that further tests be made of open hearth fumes before these estimates be widely adopted.

Region 3 has the smallest geographical area in relation to the emission of silver in that area (123,000 square miles), or in other words, the greatest emission of silver per square mile (0.88×10^{-3} tons of silver per square mile-year). Again, this is largely the result of the estimates of silver emissions from open hearth steel production.

V. NATURE OF EMISSIONS

Practically no data describing the particulate containing silver has been found. It is, therefore, necessary to estimate the characteristics of these particles based on the nature of silver and the processes that generate the emissions.

Table 4 lists some physical characteristics of silver, the most important of which may be the boiling point, 2210°C. Elemental silver, because of its high boiling point, will not be emitted in significant quantities as a vapor by processes operating at lower temperatures. Simple silver salts (halides, sulphides) and oxides are the most probable forms of silver.

TABLE 4

PROPERTIES OF METALLIC SILVER*

Melting point:	961°C
Boiling point:	2210°C
Density:	10.5 grams/cm^3
Atomic weight:	108 awu
Heat of vaporization:	60.7 kg-cal/g-atom
Hardness:	2.8 (standard minerology scale)

*Reference 30, Table 3-169. See also Table 3-1.

Whether these compound forms are produced from elemental silver at high temperatures, or the reverse, depends on the excess oxygen, sulphur, chlorine, etc., and also on the presence of cyanide ions.

Iron and steel producing processes are typically on the order of 1000°C, although in an electric furnace local temperatures must be extremely high due to the concentrated electric current. Particulates from these processes range in size from submicrometer to 5 micrometers.

There is no data to indicate the size of the silver-containing particles within this range. After release from a stack, any of these particles are capable of traveling long distances before settling to earth.

Combustion processes (coal, oil, or incineration) tend to be at lower temperatures. The particulate produced, however, is somewhat larger, ranging up to approximately 25 microns. Although the largest particles will settle out fairly quickly, smaller particles including a small fraction of submicrometer ones will remain airborne for long periods.

Since metallic silver is a relatively stable material, there is no reason to expect significant chemical changes following emission. Likewise there is no reason to expect unusual physical behavior of particles containing silver. Dispersion, agglomeration, and settling may be presumed to proceed at normal rates for these particles.

Silver is not widely known as a toxic material. In nitrate form it is applied to the eyes of babies, and is used externally in cases of severe burn. In the vicinity of non-ferrous smelters where the toxic effects of zinc and lead are well known, emissions of silver have not been reported as problematic. The extent to which silver, as an air pollutant, might be considered hazardous has not yet been established.

VI. UPDATING OF EMISSIONS ESTIMATES

The following recommendations are made for periodically updating the estimates made in this study.

A. VERIFICATION OF CURRENT ESTIMATES

1. Because the estimated high emissions levels from open hearth steel production significantly influence the conclusions of this report, improved data for this emissions source should be obtained.

2. Within the silver industry, the emission of silver from non-ferrous smelters and refineries constitutes the most concentrated source, i.e., significant emissions from a small number of sources. Therefore, within the industry, these sources offer the most obvious opportunities for improved emission control. Compilation of additional data is recommended to supplement the estimated emissions data from these sources.

B. PERIODIC REVIEW OF ESTIMATES

1. The Bureau of Mines' estimates for material flow, industry practices, and trends provide the best estimates of the size of the industry.

2. EPA activities are currently generating the best emissions data and should be reviewed using:

   a. Overall industry studies; e.g., Reference 31;

   b. The Source Test Program in which specific individual plant emissions are measured. This information provides emission factors for specific examples of typical industry operations; and also provides some analyses of the particulate, usually including trace metal content and particle size;

   c. NEDS (National Emissions Data System) is steadily being enlarged and improved. This system can provide emission factors for specific plants and plant operations, the type of particulate control equipment in use, and the actual, or estimated, control efficiency. The system may eventually be expanded to include description of the emissions.

3. The silver industry should be consulted for its opinion and suggestions regarding the most recently published estimates. This may be best accomplished by interviewing the Silver Commodity Specialist, Division of Non-ferrous Metals, Bureau of Mines in Washington; or by interviewing one or more of the principal companies in the industry.

4. The literature should be reviewed using (a) industrial views as published from time to time in Chemical Engineering, for example, and (b) environmental views as summarized in Pollution Abstracts, for example.

5. Individual companies or plants may be approached for opinions, data, or cooperative tests of their own operations. This is a difficult approach to the obtaining of fresh information due to the natural reluctance of the plants to discuss environmental problems. However, data thus obtained can have a relatively high degree of reliability.

6. State agencies in which specific plants are located may be able to provide useful information and should be contacted.

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